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Expansive admixtures (mainly ettringite)

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Abstract

The hydration chemistry of expansive cements is described with emphasis on the components for formation of ettringites. Expansive cement concrete as 'shrinkage-compensating concrete' or 'chemical prestressing concrete' is applied to many kinds of concrete construction. This paper outlines the chemical composition of calcium sulfoaluminate and lime-based expansive admixtures and discusses the expanding mechanism, chemical prestressing, and typical properties of expansive cement concrete. Finally, research and development are introduced with respect to expansive admixtures. Furthermore, these expansive admixtures are based on cement minerals, except 'gas forming admixture' and 'admixtures containing granulated iron filings'. © 1998 Elsevier Science Ltd. All rights reserved.

Keywords: concrete, expansive admixture, ettringite, shrinkage-compensating concrete, chemical prestressing concrete, expanding mechanism, expansion rate, strength, length change, durability.

INTRODUCTION

Conrete has two inherent properties such as 'shrinkage due to drying' and 'smaller tensile strength compared with compressive strength'. Consequently, it is unavoidable for concrete in a structure to crack due to drying shrinkage. These cracks are mentioned as the largest drawback of concrete structure, as they lower the durability due to the water entering to hasten the corrosion of steel bars, allow the leakage of rain, and spoil the beauty.

To cover these faults of concrete structure, study into the expansive cement has been made in foreign countries which utilizes the expansibility of ettringite. Ettringite has been detested as 'a cement-bacillus' which causes expansive fracture of concrete. In 1936, Lossier[1] began a study to produce chemically prestressed concrete and thereafter Lafuma[2] and Klein[3] took over the study to build up a foundation of the expansive cement.

ACI Standards (proposal)[4] show three kinds of expansive cement, that is, 'K type', 'M type' and 'S type'. In Japan, the beginning of the growth of expansive concrete was felt in the mid-1960s with the development of expansive admixtures. Since then for more than 20 years not only the basic research but also the investigation into practical use has been actively continued. Expansive concretes developed in Japan, unlike those in America and Europe which have been made with the use of expansive cement, are manufactured by mixing expansive admixtures with concrete, with the result that control and management of expansion rate are made easy. Therefore, expansive concretes of Japanese type are widely used not only in shrinkage-compensating concrete but also in chemical prestressed concrete which positively utilizes the expansibility of them. Production of expansive admixtures in Japan is roughly estimated at 70 000 metric tons in 1995. Therefore, about 1 700 000 m^{-3} of expansive concrete was used yearly. Recent tendency toward reconsideration of cracks in concrete structures from the viewpoint of durability may have created a new need for the expansive admixture as a method of crack prevention. Moreover, a certain type of expansive admixture, with a property to control the 'the thermal cracking due to the heat of hydration of cement' which has a possibility to be caused with an increase of larger concrete structures, has also been put to practical use.

In this paper, properties of expansive admixture and concrete are expressed and also the present technical problems and the trend of development are stated.

PROPERTIES OF EXPANSIVE ADMIXTURES

Expansive admixtures have such as iron powder, alumina powder, magnesia, calcium sulfo aluminate (CaO–Al_3O_2–SO_3) and calcium oxide (CaO). However, the main groups are the calcium sulfo aluminate series.

ACI STANDARDS (proposal) in the USA[4] include three types of expansive cement as follows:

(1) K-type: portland cement mixed with anhydrous Hauyne ($3CaO{\cdot}3Al_2O_3{\cdot}CaSO_4$), gypsum ($CaSO_4$) and quick lime ($CaO$).
(2) M-type: portland cement mixed with alumina cement and gypsum ($CaSO_4$) at a reasonable ratio.
(3) S-type: normal portland cement mixed with larger amount of tricalcium aluminate (C_3A) and gypsum ($CaSO_4$).

Figure 1 shows a three-component system[5] of an expansive admixture relating to calcium sulfo aluminate series. When a component including these minerals hydrates, various kinds of hydrates are produced. Of these hydrates the

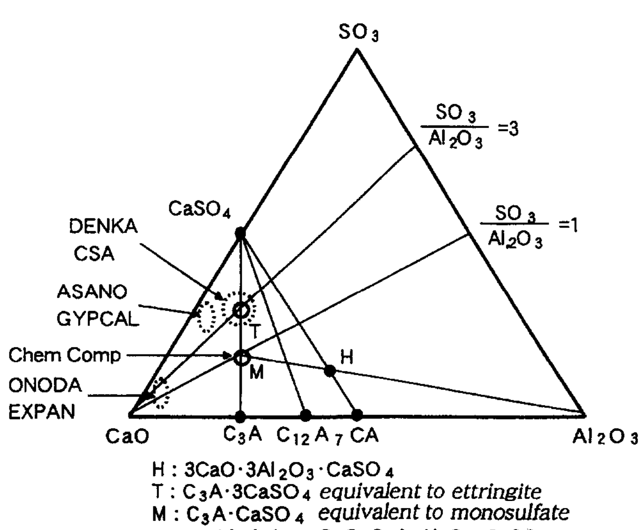

Fig. 1. Diagram of expansive admixtures.

followings are mentioned as expansive; ettringite ($C_3A{\cdot}3CaSO_4{\cdot}32H_2O$) shown at T-point, monosulfate ($C_3A{\cdot}CaSO_4{\cdot}12H_2O$) shown at M-point and calcium hydroxide ($Ca(OH)_2$) shown on CaO.

Examples of calculation for the volume change at the time of hydration of expansive hydrate are shown as follows:

Production of ettringite

$$C_4A_3S + 8CaSO_4 + 6CaO + 96H_2O \rightarrow 3C_3A{\cdot}3CaSO_4{\cdot}32H_2O$$

	C_4A_3S	$8CaSO_4$	$6CaO$	$96H_2O$		$3C_3A{\cdot}3CaSO_4{\cdot}32H_2O$
Weight	610	1088	336	1728		3762
Specific gravity	2·61	2·96	3·34	1		1·73
Volume	(234	368	101	1728)	[2431]	2175

Rate of volume change for hydrates series 2175–2431/2431 × 100 = −10·5%

Ratio of volume change for C_4A_3S particle 2175/234 = 9·3 times

Production of monosulfate

$$C_4A_3S + 2CaSO_4 + 6CaO + 36H_2O \rightarrow 3C_3A{\cdot}CaSO_4{\cdot}12H_2O$$

	C_4A_3S	$2CaSO_4$	$6CaO$	$36H_2O$		$3C_3A{\cdot}CaSO_4{\cdot}12H_2O$
Weight	610	272	336	648		1866
Specific gravity	2·61	2·96	3·34	1		1·95
Volume	(234	92	101	648)	[1075]	957

Rate of volume change for hydrates series 957–1075/1075 × 100 = −10·9%

Ratio of volume change for C_4A_3S particle 957/234 = 4·1 times

Production of $Ca(OH)_2$

$$CaO + H_2O \rightarrow Ca(OH)_2$$

	CaO	H_2O		$Ca(OH)_2$
Weight	56	18		74
Specific gravity	3·34	1		2·24
Volume	(17	18)	[35]	33

Rate of volume change for hydrates series 33–35/35 × 100 = −5·7%
Ratio of volume change for CaO particle 33/17 = 1·9 times

Several expansion mechanism of expansive cement concrete have been proposed as follows:

(1) volume increase of gel state expansive ingredients by water adsorption (swelling theory)[6,7]
(2) spreading out of surroundings by crystal growth of crystalline expansive ingredients (crystal growth theory)[8–11]
(3) forming coexisting pores by disintegration of expansive ingredients during hydration[12]

In either case, 'formation of pores in the hardened structure' or 'formation of gel state hydrates with low density' is required for the 'expansion' to coexist with 'chemical shrinkage'. Further studies are needed to examine quantitatively the formation of pores and gel state hydrates, and to discuss including the chemical shrinkage and autogenous shrinkage[13] (or autogenous expansion). In the case of expansion taking place with formation of ettringite or CH, topo-chemical reaction at the surface of expansive ingredients is widely accepted rather than through-solution reaction. The important factor in expansion is not merely the hydration of expansive ingredients, but the formation of surrounding hydrates which the compressive force produced from the expansive ingredients are transmitted to.[14] Namely, expansion does not take place otherwise the hardened matrix structure is formed by hydration of cement. Accordingly, it is also importnat that the hydration of both expansive admixtures and cement must take place at the opportune moment.

Japanese definition of expansive admixture explains that 'Admixtures, when mixed with cement and water, produce ettringite ($3CaO \cdot Al_2O_3CaSO_4 \cdot 32H_2O$) or calcium hydroxide ($Ca(OH)_2$) by hydration reaction to expand the concrete'. Table 1 shows the physical properties and chemical compositions of both 'ETTRINGITE series' and 'LIME series'. Figure 2 shows their chemical reaction formula. Table 2 shows the quality standards prescribed in JIS A 6202 'Expansive Admixtures for Concrete' which are now under revision to secure unity with 'ISO Standards'.

Table 1. Physical properties and chemical composition

Item		*C–S–A type*	*CaO type*
Physical properties	Specific gravity (%)	3·00	3·14
	Specific surface area (cm^2/g)	2500	3500
Chemical composition	Ignition loss (%)	0·8	0·4
	SiO_2 (%)	4·0	9·6
	Al_2O_3 (%)	10·0	2·5
	Fe_2O_3 (%)	1·0	1·3
	CaO (%)	51·2	67·3
	MgO (%)	0·6	0·4
	SO_3 (%)	31·9	18·0
	Total (%)	99·5	99·5

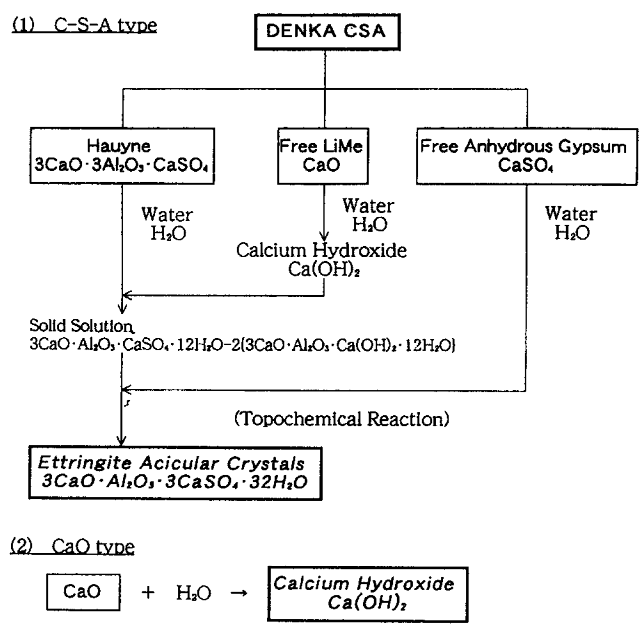

Fig. 2. Schematic representation of the reaction.

Table 2. Specifications of expansive admixtures

Items			*Specified value*
Chemical composition	Magnesium oxide (%)		5·0 max
	Ignition loss (%)		3·0 max
Physical properties	Specific surface area (cm^2/g) .		2000 min
	Residue on 1·2 mm sieve* (%)		0·5 max
	Set: Initial setting (min)		60 and after
	Set: Closing of setting (h)		Within 10
	Expansivity (rate of length change)	7 days	0·000 30 min
		28 days	−0.000 20 min
		3 days	6·9 min
	Compressive strength (MPa)	7 days	14·7 min
		28 days	29·4 min

*The 1·2 mm sieve is 1190 μm aperture standard sieve specified in JIS Z 8801.

PROPERTIES OF EXPANSIVE CONCRETE

When expansive concretes are classified based on size of expansive force, they may be broadly divided into shrinkage compensating concrete and chemical prestressing concrete.

Fresh concrete of expansive concrete shows about the same nature with ordinary concrete in which expansive admixtures are absent.[15]

Figure 3 shows the expansion rate of the expansive concrete in the relation between unit expansive admixture content and nonrestraint expansion rate.[16] It shows that the expansive rate becomes larger with the increase of unit expansive admixture content having good relation with unit expansive admixture content. Figure 4 is an example showing relations among unit expansive admixture content, compressive strength and expansion rate.[17] It is shown that restraint expansion rate becomes larger with the increase of unit expansive admixture content. However, the compressive strength of expansive concrete having unit expansive admixture content not much exceeding 30 kg/m^3 and cured under nonrestraint condition shows about the same value with ordinary concrete. The compressive strength decreases when expansive rate increases to some extent. It is possible that when the expansive concrete is allowed to expand without restraint the structure of the concrete becomes loosened to cause the lowering of strength. However, the expansive concrete allowed to expand under the restrain

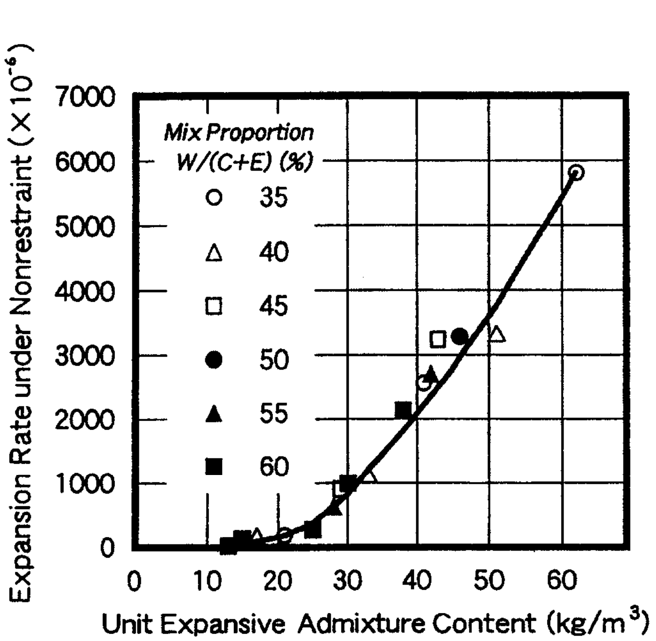

Fig. 3. Relation between unit expansive admixture content and nonrestraint expansion rate.

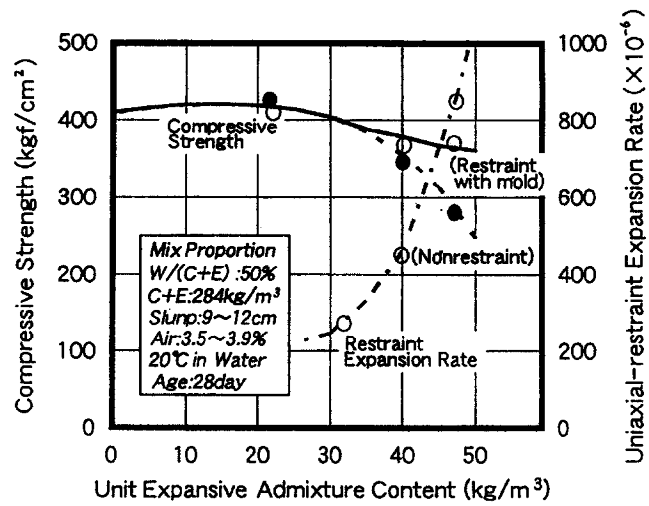

Fig. 4. Relations among unit expansive admixture content, compressive strength and restraint expansion rate.

of steel mold does not show such lowering of strength and shows about the same strength as the ordinary concrete, irrespective of large expansive rate. It is possible that in the latter case the structure of concrete is kept compact by restraining to cause no lowering of strength.

Under the condition where unit expansive admixture content is not much exceeding 30 kg/m^{-3}, tensile strength, bending strength, bond strength, Young's modulus and creep of the expansive concrete are about the same as ordinary concrete.[15]

As for durability, the expansive concrete given long time water curing does not change the expansive rate after it has reached a settled value. Though the expansive concrete dry cured after water curing shows shrinkage due to drying, it is generally recognized that the shrinkage rate is smaller than ordinary concrete. One of the examples is shown in Fig. 5.[18] When unit expansive admixture content not much exceed-

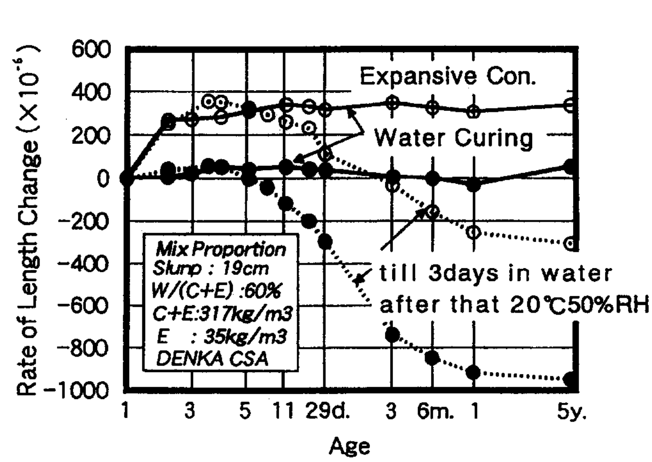

Fig. 5. Rate of length change.

ing 30 kg/m^{-3} are air entraining agent is used, freezing and thawing resistance shows about the same value as the ordinary concrete. However, when unit expansive admixture content not much exceeding 50 kg/m^{-3} is used and concrete is allowed to expand with unsatisfactory restraining, it is reported that frost damage resistance is sometimes lowered.[15] The element which exerts the greatest influence on durability factor is the kind of supplementary AE agent. Therefore, it is shown that the durability can be improved by increasing the air volume and restraining with reinforcing bars, even if unit expansive admixture content is not less than 45 kg/m^{-3}.[19]

Figure 6[19] shows the relation between durability factor and boid distance factor in expansive concrete. It shows that durability factor of steel fiber reinforced expansive concrete gives little change with the variation of boid distance factor. As for the expansive concrete without steel fiber durability factor shows great fall when the boid distance factor takes not less than 300 μm, with the result that the same relation as ordinary concrete between durability factor and boid distance factor is formed.

With regard to comparison of samples of expansive concrete and ordinary concrete taken from actual concrete structures of 22 years old, expansive concrete and ordinary concrete showed 28 day compressive strength of 18·6 and 18·1 MPa respectively, that is, almost the same strength; after 8 years, 56·3 MPa of compressive strength; after 22 years, 59·0 MPa of compressive strength; after 22 years of exposure, samples taken from the actual concrete structures showed satisfactory compressive strength (Fig. 7).[20] Figure 8[20] shows the carbonation depth measured on the cut surface of core test

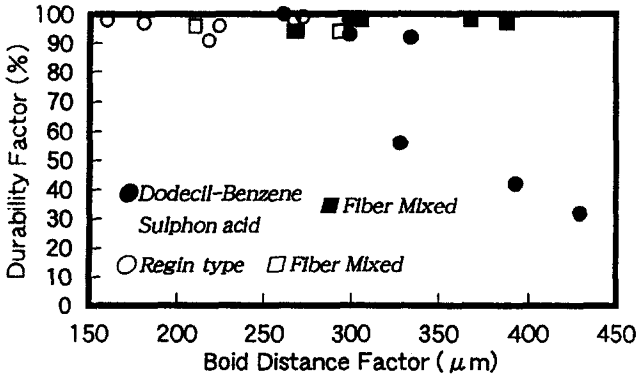

Fig. 6. Relation between boid distance factor and durability factor.

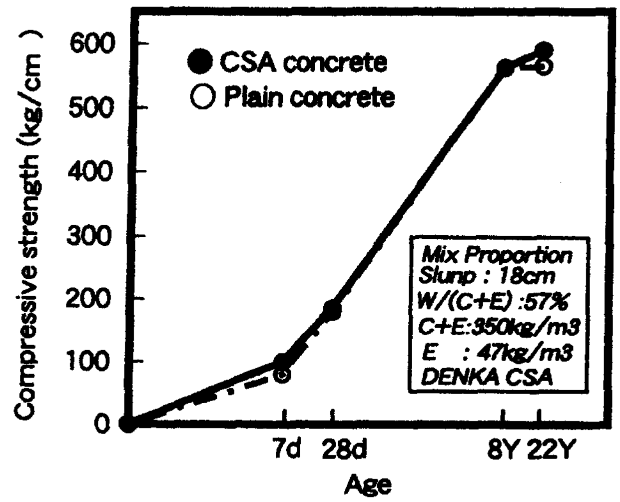

Fig. 7. Compressive strength of concrete.

specimens applied with phenolphthale in which had been exposed for 8 and 22 years respectively. Results of measurement of carbonation depth by phenolphthale in method gave about the same value.

Though it is reported that sulfate resistance and wear resistance of expansive concrete show about the same values as ordinary concrete, it is recognized that water tightness of expansive concrete cured under restrained condition shows a tendency to give smaller value than ordinary concrete.[21] This may be due to the compact structure of the expansive concrete resulted from compressed air void in the concrete by press effect owing to the restraining of concrete during curing (Fig. 9).

Expansive concrete has a chemical prestress effect which introduces compressive stress into concrete by restraining the expansive energy

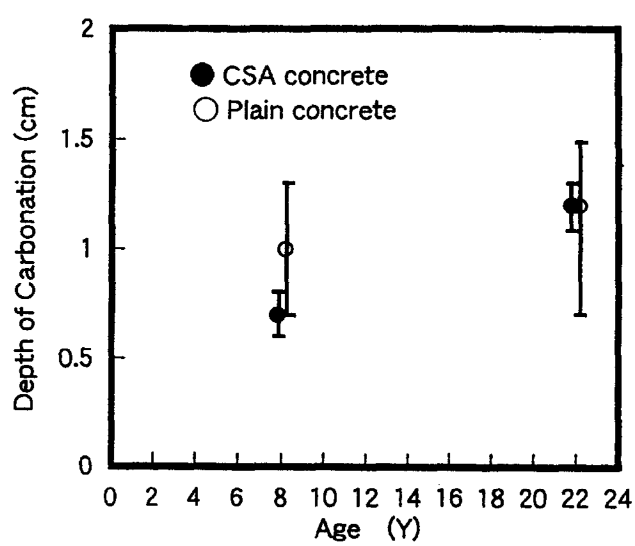

Fig. 8. Carbonation depth of concrete.

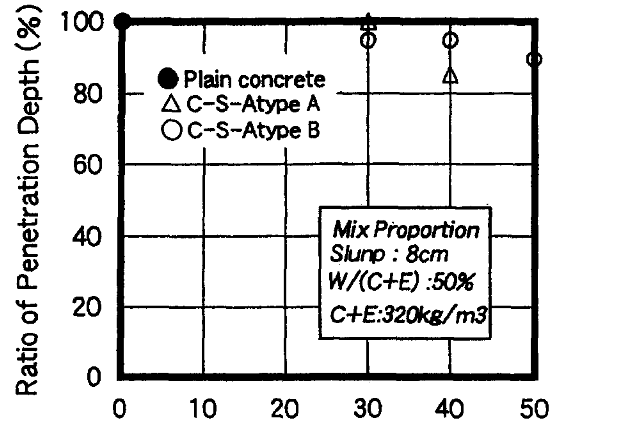

Fig. 9. Result of permeability test.

produced from hydration reaction. However, the chemical prestress varies greatly in accordance with such factors concerning mix proportion of concrete as unit expansive admixture content, kind of cement and unit binder content, water–binder ratio, and curing method and is also highly influenced by the amount of restrained and restraining method. As for the restrained, reinforcing bar, structural steel, steel pipe, ground and existing concrete structure are thinkable. Therefore, it is necessary to assume the chemical prestress for the arrangement of these restrainers and their amount. After all, to utilize the expansive energy it is the most important point to establish the estimating method for it. As for the chemical prestress under unit-axial restraining condition, the method of test is prescribed in JIS A 6202. And according to the method basing on 'the notion concerning amount of work,[22] the assuming method of chemical prestress can be calculated by the following equation.

$$U = \varepsilon_x \cdot \sigma_{cpx} \qquad (1)$$

where:

U amount of work done to a restrainer by unit volume of expansive concrete
ε_x expansion rate
σ_{cpx} chemical prestress

Generally the amount of work U can be assumed as constant within the limits of practical restraint.

Using expansive coefficient ε_{ss} obtained from a restrained specimen according to A-method or B-method prescribed in JIS A 6202, and amount of work U calculated from following equation, chemical prestress σ_{cpx} in the above-mentioned eqn 1 can be computed.

$$U_s = p_s \cdot E_s \cdot \varepsilon_{ss}^2 \qquad (2)$$

where:

p_s restraining steel ratio of uniaxially restrained standard specimen (= 0·96%)
E_s Young's modulus of restraining steel

However, in the actual concrete structure it is unusual to have a restrainer only in one direction. For example, reinforcing bars in slab are arranged in two directions and in beam and column members it is arranged in three directions. Generally the expansive concrete has multiaxial restraint such as biaxial or triaxial restraint. There are some reports discussing the expansive properties, compressive strength, test method of expansive amount and assuming method of expansive amount, of expansive concrete which has multiaxial restraint.[23,24] As for the expansive concrete writers proposed that it is a question to consider the expansive properties of concrete under multiaxial restraint simply and easily as an amplification of uniaxially restrained expansion and also offered that the expansive properties with annular restraint can be assumed by multiaxially amplify 'the notion concerning amount of work' in uniaxial restraint.[25]

EXAMPLES OF USE AND FUTURE VIEW OF EXPANSIVE CONCRETE

As for the records of performance of expansive concrete, the following may be mentioned.

(1) Application to shrinkage-compensating concrete[26] for cast-in-place concrete.
(2) Application to shrinkage-compensating concrete of factory products and to chemical prestressing concrete.[26]
(3) Application to filling concrete for placement in inner space formed by existing concrete, steel pipe and rock.

As stated before yearly production of expansive concrete in Japan was 70000 metric tons in 1995. And the share of application was 30% for cast-in-place concrete and 70% for precast concrete.

As for the use of cast-in-place concrete, water utilizing structures such as water tank, underground pit and pool, and structures such as bridge slab and pavement slab, and watertight concrete may be mentioned. In recent years, with a tendency of concrete structure to grow larger, also the expansive admixture of hydration heat reducing type[27] which has an addition of hydration heat reducing admixture is used.

Concrete structures with comparatively large section (mass concrete) in such facilities as water treatment installation tend to cause thermal cracks due to the hydration heat of cement instead of shrinkage cracks in smaller structures. The object of the use of expansive admixture of hydration heat reducing type is to reduce the hydration heat of cement and to decrease the thermal stress by relaxing the restraining stress during the drop of temperature with expansive energy maintaining. Results of execution test by actual structures on a job site show that with the use of expansive admixture of hydration heat reducing type it is recognized that width and number of crack is diminished and also a part of crack disappear with the lapse of time showing the recovering effect of expansive action,[28] though the complete prevention of thermal cracks is not yet achieved.

It was also considered that the use of expansive concrete in place of reinforced concrete slab of highway steel bridge may have effect on diminishing of crack due to the introduction of chemical prestress and shrinkage decreasing effect.[29] They made a trial execution on actual, bridge and recognized that the results of investigation and measurement proved the decrease of crack to improve durability.[30–34]

As for the use of precast concrete, Hume pipe, box culvert, precast slab and steel pipe lining are mentioned. These are the products which positively use the expansive admixture to introduce chemical prestress. Introduction of chemical prestress into concrete is achieved by restraining the expansive force of expansive concrete with reinforcing bars to obtain reaction. With this method it is aimed to increase the crack resistance or to diminish the sectional area of member.

In addition to the revision of JIS STANDARD for Hume pipe, under the confirmation of Japan Institute of Construction Engineering Box Culvert Association enacted 'Guideline for Buried Box Culvert Made of Precast Reinforced Concrete' which classified the products into two classes, that is, the First class and Second class according to the resistivity to external pressure. With these efforts, concrete product having chemical prestress or expansive admixture presents ever-increasing demand.

In recent years with the object of (1) labor saving of concreting works, (2) complete filling of concrete to the structure with complex sectional shape or high density re-bar arrangement, (3) arresting of vibration and noise to the neighborhood, (4) improving of high strength concrete execution, high fluidity concrete[35,36] has been applied to the actual works on a job site to increase performance records. Though sometimes cracks appear in a high fluidity concrete applied to a large-scale project, it may be due to the mix proportion of the high fluidity concrete in which the proportion of powders such as cement, iron-blast-furnace slag, fly ash and powdered lime stone increase, with the result that thermal shrinkage due to the hydration heat of cement, drying shrinkage and 'Autogeneous-shrinkage'[37] of cement paste due to the hydration reaction become large. With the object of compensating the shrinkage of high fluidity concrete, a high fluidity concrete with expansive admixture is used in a wall concrete of installation such as water treatment facilities where the members have a thickness of over 1 m. Result of comparison between the high fluidity concrete with expansive admixture and the high fluidity concrete without expansive admixture shows that the former presents a slower rate of increase in the initial tensile strength with the lapse of age than the latter, with the result that the former has low incidence of crack. This also proves the shrinkage compensating effect of expansive admixture in a high fluidity concrete.

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